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IRRADIATION BEHAVIOR OF RESTRAINED AND VENTED URANIUM-2 w/o ZIRCONIUM ALLOY

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ABSTRACT

Twelve 0.22-in.-diameter fuel specimens containing a longitudinal central vent and clad with 0.010 in. of Type 304 stainless steel were irradiated to evaluate the effect of restraint and a central vent on fuel element stability. The cladding of 10 of the specimens contained porous end plugs to vent any released fission gas and thus to minimize the buildup of gas pressure within the stainless steel cladding. The specimens consisted of a 20 per cent enriched uranium-2 w/o zirconium alloy core surrounded by a natural uranium-2 w/o zirconium alloy sleeve. This geometry was employed to produce a radial temperature gradient similar to that which will exist in the fuel of the fast breeder reactor EBR-II. The central vent was provided to enhance the release of fission gas and other volatile fission products from the fuel, thereby reducing the tendency for swelling. Eight of the specimens were irradiated to burnups of the enriched core of 6.9 to 12.8 per cent of all atoms (1.2 to 2.2 a/o of the duplex assembly) at maximum fuel temperatures ranging from 280 to 760°C. Most of the clad specimens exhibited negligible volume increases as a result of irradiation. Two specimens containing central vents but unclad were irradiated together with the clad specimens in an attempt to differentiate between the effects due to a central vent and the effects due to cladding. The central vent in itself did not appear to reduce the swelling characteristics of the alloy. Mechanical restraint appeared to have extended the useful operating temperatures of the metallic fuel alloy by at least 200°C and also greatly extended the burnup levels to which the fuel could be irradiated. This approach is of greatest interest for fast reactors, which can more readily tolerate the amounts of cladding necessary to provide restraint, and in which high burnups, high fuel temperatures, and high fissile and fertile atom densities are required.

INTRODUCTION

Since single crystals of uranium, as well as polycrystalline uranium, exhibit anisotropic growth under neutron irradiation,[1] the best that can be expected for polycrystalline material are the properties of a randomly oriented structure. Therefore, to achieve dimensional stability in uranium and uranium-base alloy fuel elements, a promising approach would seem to be to rely on the restraining influence of strong cladding materials. Although the cladding on many reactor fuel elements is known to assist in maintaining dimensional stability of the elements under irradiation, there is little quantitative information available on the irradiation behavior of uranium and uranium alloys under restraint, especially of uranium alloys irradiated to several atom per cent burnup at maximum fuel temperatures above 600°C.

At the time (1954) this investigation was initiated, the following facts were known concerning irradiation of uranium and uranium-base alloys under restraint. In the first core of EBR-I, Type 347 stainless steel cladding, 0.020 in. thick, was able to restrain the anisotropic growth of 0.35-in.-diameter beta-quenched uranium at a maximum fuel temperature of 400°C at burnup levels of 0.25 a/o.[2] In an experimental uranium rod, 0.050 in. in diameter, growth was suppressed by 0.006 in. of Type 347 stainless steel cladding to burnup levels of 1.0 to 1.2 a/o at a maximum fuel temperature of approximately 400°C.[3,4] Other early work at Argonne National Laboratory indicated that 0.010 in. of Type 304 stainless steel, Globe iron, or zirconium was able to partially suppress surface roughening in cast uranium-5 w/o chromium alloy which had a large grain size.[5] The irradiations were performed at temperatures greater than 600°C to a burnup of approximately 0.25 a/o. In these experiments the stainless steel was the most effective; zirconium was least effective, and Globe iron was intermediate between the two. Eichenberg at WAPD had also reported that 0.020 in. of zirconium cladding was not sufficient to restrain the anisotropic growth of alpha-rolled and swaged natural uranium at irradiation temperatures between 70 and 140°C.[6] The burnup levels in his experiment were not reported but are assumed to be low, since the total irradiation time was only 6 weeks.

In addition to these irradiation experiments, Smith, Zegler, and Mayfield at Argonne studied the ability of zirconium, Type 347 stainless steel, and tantalum to suppress the thermal-cycling-induced growth of uranium.[7] The lower cycling temperatures were 100 or 150°C, and the upper cycling temperatures ranged from 500 to 800°C. Zirconium in any of the thicknesses tried was ineffective in suppressing growth at temperatures above 550°C. For equal cladding thickness, tantalum was more effective in suppressing growth than was Type 347 stainless steel for temperatures up to 800°C.

As can be seen, most of the previous information dealt with the problem of anisotropic growth in wrought material and surface roughening in coarse-grained cast material. Today, with the greater emphasis on achieving high temperatures in operating reactor fuel elements, irradiation-induced swelling of metal fuels is a problem of significantly greater importance than either anisotropic growth or surface roughening. The latter 2 problems can now be effectively minimized by suitable alloying, fabrication, and heat treatment. Also, in most metal fuels that operate at temperatures greater than about 450°C, swelling replaces anisotropic growth as the predominant mechanism of dimensional instability.

Two irradiation experiments performed at Argonne National Laboratory in 1954 and 1955 led to the development of the vented and clad fuel assembly as a means of improving the capabilities for high-temperature performance of metal fuel.

In the first experiment, the dimensional stability of restrained versus unrestrained uranium was evaluated. The specimen material was 10 per cent enriched uranium and consisted of 4 cast and five 300°C swaged and beta-quenched specimens, 0.165 in. in diameter and 1.000 in. long. The cast material was coarse grained and randomly oriented; under irradiation it could be expected to develop severe surface roughening. The wrought material was fine grained but contained some residual orientation so that anisotropic growth would normally occur during irradiation.

All but one of the specimens in each of the 2 groups were clad with Type 304 stainless steel jackets, as shown in Figure 1. Each jacketed specimen was first wrapped in 0.001-in.-thick zirconium foil to prevent diffusion or eutectic formation between the uranium and the stainless steel jacket. The radial clearance between the specimen and its cladding was 0.005 in., part of which was taken up by the foil. The jackets were used with 2 wall thicknesses, 0.010 and 0.020 in. In the shoulders of each jacket were placed holes to permit entry of surrounding NaK into the clad and foil enclosures. Set screws were placed in each end of the jacket and run down tightly against the ends of the enclosed specimen.

The assembled specimens, along with one bare control specimen from each group, were irradiated in the MTR in NaK-filled capsules to burnups of approximately 1.0 atom per cent. A summary of the irradiation conditions and observed changes is given in Table I. The calculated temperatures, based on flux measurements made with attached cobalt monitors, ranged from 670 to 910°C in the specimens and from 490 to 630°C in the jackets. A typical clad specimen after irradiation is shown in Figure 2.

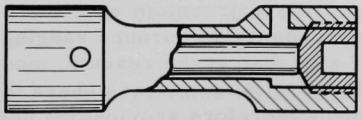


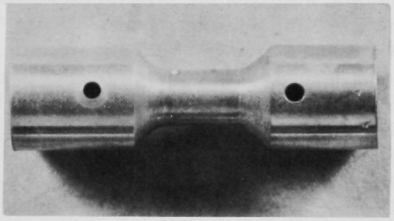
Figure 1. Uranium Specimen Jacketed in Stainless Steel for Irradiation Tests.

Table I

EFFECT OF IRRADIATION ON UNALLOYED URANIUM SPECIMENS CLAD WITH TYPE 304 STAINLESS STEEL

Specimen No.	Specimen History	Cladding Thickness, in.	Burnup, a/o	Maximum Specimen Temp, °C	Clad Temp, °C	Length Change, %	Diameter Change, %
G-4	Cast	0.010	1.0	850	620	-0.16	0.51
G-5	Cast	0.020	1.0	910	630	-0.55	0.25
G-6	Cast	0.010	1.0	860	620	-0.55	0.51
G-16	Cast	Unclad	1.0	790	-	(a)	(a)
G-10	Wrought	0.020	1.2	800	550	0.23	1.5
G-11	Wrought	0.010	1.0	670	490	-0.7	1.5
G-14	Wrought	0.020	1.0	740	510	0.16	-0.25
G-15	Wrought	0.010	1.1	730	530	0.23	1.0
G-17	Wrought	Unclad	1.2	720	-	12	(a)

(a)Measurement could not be obtained because of extreme swelling of the specimen.



16522 2X

Figure 2

Specimen G-10 after Irradiation. The Appearance of the Specimen Is Unchanged after 1.2 a/o Burnup at a Central Temperature of 800°C. The Specimen Is Clad with 0.020 in. of 304 Stainless Steel.

It was found that the dimensions of the jacketed assemblies changed only very slightly as a result of the irradiation. The cast group tended to shorten about 0.005 in., but most of the swaged and heat-treated group lengthened 0.002 or 0.003 in. The diameters of both groups increased about 0.002 in. In contrast, the bare control specimens, examples of which are shown in Figures 3 and 4, were highly deformed. Both bare specimens ignited and burned to oxide during the hot-cell examination. It was concluded from these preliminary tests that both the 0.010-in. and 0.020-in. cladding were effective in restraining anisotropic growth and distortion in the uranium, at least in the temperature range and clad-to-fuel ratios that were used.

In the second experiment, 9 uranium-2 w/o zirconium specimens containing a longitudinal central vent were irradiated to burnups ranging from 2 to 3 a/o. Unfortunately, because of a mishap at the reactor, most of the specimens were inadvertently irradiated at temperatures above the melting point of the material; these specimens therefore provided no additional information. However, the information obtained from the few specimens which operated below the melting point indicated that the central vent may have enhanced the ability of the fuel to resist high-temperature swelling

16437 2X

Figure 3. Cast Unclad Specimen G-16 after 1.0 a/o Burnup at 790°C. The Right End of the Specimen Ignited During Photographic Exposure.

16526 2X

Figure 4. Wrought Unclad Specimen G-17 after 1.2 a/o Burnup at 720°C. The Left End of the Specimen Ignited Shortly after the Specimen Was Photographed.

On the basis of these results, an irradiation experiment involving a dual approach to minimize high-temperature swelling in metal fuels was proposed by F. G. Foote. Since specimens of uranium and uranium-base alloy fuels had been shown to develop large central voids when irradiated with very high radial temperature gradients and at central temperatures above 600°C, presumably due to fission gas agglomeration,[8] a 0.031-in.-diameter, full-length axial hole was to be provided in each fuel specimen. The hole was intended to provide an effective escape route for fission gases migrating to the center of the fuel. Escape of the fission gas from the specimen would reduce the pressure due to the agglomeration of fission gases in the center of the specimen, thereby reducing the tendency for central void formation. Next, the fuel was to be clad with a material possessing sufficient mechanical strength to restrain swelling at the temperatures and burnups of interest. The cladding was to contain porous end caps to enable gaseous fission products to be released from the fuel assembly, thereby lowering the pressure within the cladding. In addition, the porous end caps were intended to provide information on the diffusion of gaseous and solid fission products through various types of porous barriers into the surrounding NaK of the irradiation capsule.

This report contains the information obtained by the irradiation of 14 such vented and clad fuel assemblies. The irradiations were made in the MTR during 1957 and 1958.

SPECIMEN MATERIAL AND PREPARATION OF IRRADIATION SPECIMENS

Considerable development work had been done at Argonne National Laboratory on the fuel alloy uranium-2 w/o zirconium. Earlier irradiation studies[9] had shown good irradiation stability for this alloy at temperatures below 350°C, as shown in Figure 5. However, the material

undergoes large volume increases when irradiated at temperatures above 600°C, as shown in Figure 6. In conjunction with the large amount of swelling observed in specimens irradiated at high temperatures and with high radial temperature gradients, the specimens also developed longitudinal central voids, such as that shown in Figure 7. Since considerable knowledge had been obtained for unclad uranium-2 w/o zirconium alloy, this alloy was selected for the high-temperature, high-burnup irradiation studies with vented and clad fuel assemblies.

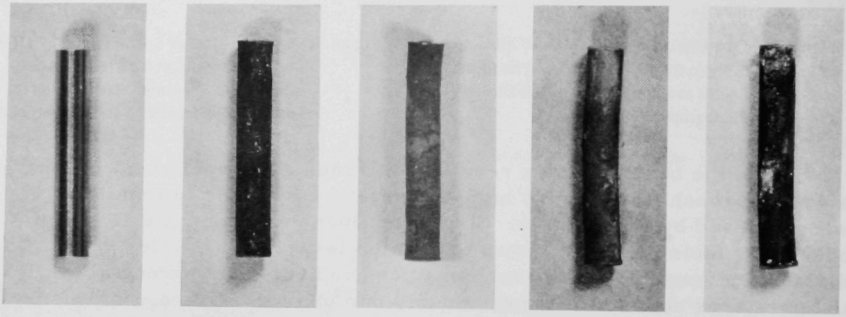


Figure 5. Effect of Irradiation below 350°C on Cast Uranium-1.62 w/o Zirconium Alloy.(9)

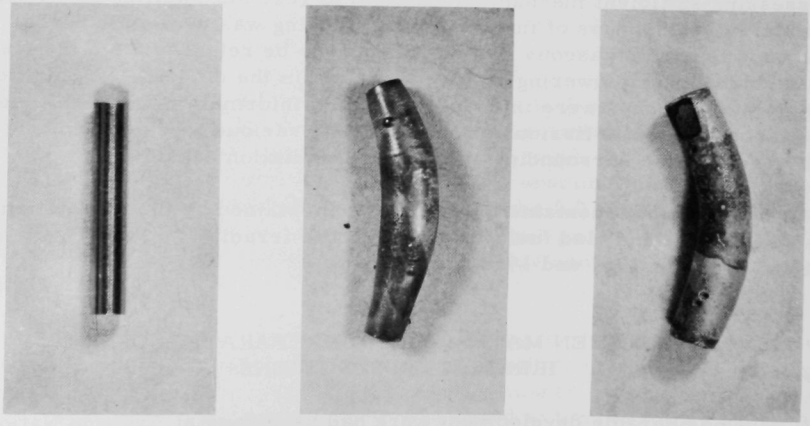
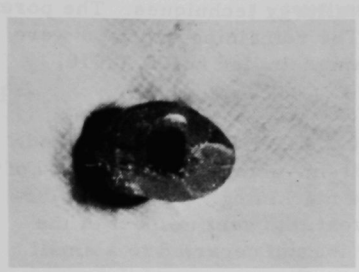


Figure 6. Effect of Irradiation above 600°C on Cast Uranium-1.62 w/o Zirconium Alloy.(9)



106-3431 4X

Figure 7. Central Void in Uranium-1.62 w/o Zirconium Alloy Specimen after 5.2 a/o Burnup at 700°C.(9)

A schematic view of the vented and clad fuel assembly is shown in Figure 8. The assembly consisted of a central core of uranium-2 w/o zirconium alloy (20 per cent enriched), 3 in. long with an ID of 0.031 in. and an OD of 0.094 in., upon which was shrink-fitted a natural uranium-2 w/o zirconium alloy sleeve with an ID of 0.094 in. and an OD of 0.219 in. The regions of differing enrichment were intended to provide a radial temperature gradient under irradiation that would simulate that expected in the EBR-II reactor. Exposure to an unperturbed thermal neutron flux of 1 x 10^{14} neutrons/cm^2-sec was calculated to produce a maximum fuel center temperature of 850°C in the fuel assembly.

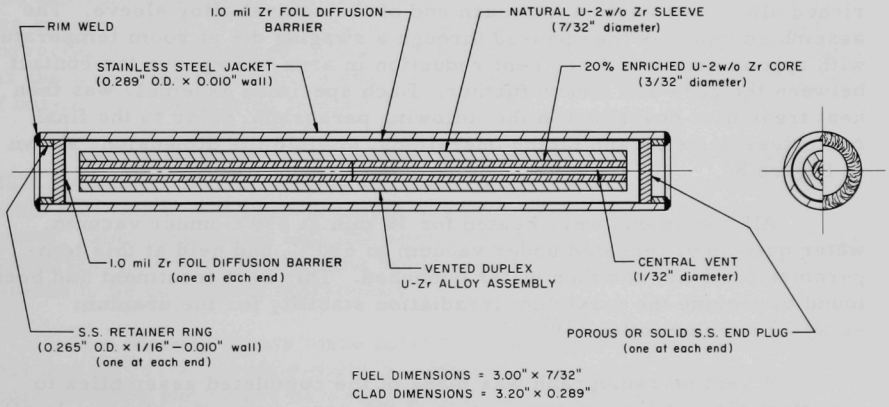


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Figure 8. Longitudinal Section of Restrained and Vented Uranium-2 w/o Zirconium Alloy Fuel Assembly.

The fuel was jacketed with 0.010-in.-thick Type 304 stainless steel cladding. Between the fuel and the cladding was placed a 0.001-in.-thick zirconium foil to prevent eutectic formation between the uranium alloy and the stainless steel. Also, there was a 0.010-in. annulus of NaK between the stainless steel and the outer surface of the natural uranium-2 w/o zirconium alloy sleeve. The porous end caps which provided endwise restraint of the fuel were cut from 0.063-in.-thick porous sheets of Type 304 stainless

steel which had been produced by powder metallurgy techniques. The pore sizes in the end caps were 5, 20, and 65 μ. The remaining end caps were 0.020 in. thick and were either solid or contained drilled holes, 0.010, 0.020, or 0.050 in. in diameter.

The enriched core was statically cast and then rolled and swaged to the final outside diameter. The hole of 0.031-in.-diameter in the center of the core was then formed by electrical discharge machining. Because of difficulties encountered in drilling the central vent, the vent in most of the specimens was larger than the desired 0.031 in. and departed to a small degree from an exactly circular cross section. The core material was cut into 1.50-in. lengths prior to boring the hole since it was impossible at that time to drill the holes the length of the entire specimen. Two 1.50-in. pieces were butted together to give a 3-in. specimen. The natural uranium-2 w/o zirconium sleeve was fabricated from a rod that had been hot rolled at 600°C. The sleeve was then centerless ground to the final outside diameter and cut into 3-in. lengths. The 0.094-in. hole in the sleeve was drilled from each end.

The specimens were assembled by quickly forcing a chilled, enriched alloy core section into each end of the natural alloy sleeve. The assembled unit was then passed through a swaging die at room temperature with approximately a 4 per cent reduction in area to improve the contact between the core and sleeve further. Each specimen assembly was then heat treated as described in the following paragraph, prior to the final centerless grinding and facing operations, to yield the dimensions shown in Figure 8.

All specimens were heated for 10 min at 850°C under vacuum, water quenched, reheated under vacuum to 690°C, and held at this temperature for 6 hr, and then water quenched. This heat treatment had been found to provide the maximum irradiation stability for the uranium -2 w/o zirconium alloy.(10)

A gamma radiograph was made of the completed assemblies to ascertain the positioning and seating of the core within the sleeve. In all cases the 2 core sections appeared to have been tightly butted against each other at the center of the sleeve.

The NaK bond between the fuel and cladding was accomplished as follows. The specimens were placed in their irradiation capsules and the capsules were evacuated. The capsules were then completely filled with NaK, closed, and heated to expand the NaK into the region between the fuel and the cladding. This operation was repeated several times until the region between the fuel and cladding was entirely filled with NaK. Upon completion of this operation, the volume of NaK in the capsules was reduced to that desired during irradiation.

EXPERIMENTAL PROCEDURES

The following properties of the clad specimens were measured prior to irradiation:

1. Dimensions, to 0.001 in.
2. Weights, to 0.1 mg.
3. Density of unclad specimens and specimens with solid end caps, to 0.05 per cent, by immersion in CCl_4.
4. Mass spectrographic analysis for uranium isotope concentration in both the enriched core and natural sleeve.

Each specimen was irradiated in the MTR in a zirconium irradiation capsule similar to that used for previous irradiations.(11) To provide an efficient method of heat transfer from the specimen to the process water flowing past the capsule, each capsule contained sufficient eutectic NaK to completely cover the specimen. Each capsule contained an aluminum-0.5 w/o cobalt-0.5 w/o manganese alloy neutron-flux monitor. The neutron-induced Co^{60} activity in the monitor was analyzed by gamma-ray spectrometry. The flux incident on each capsule was obtained after corrections for resonance activation of the cobalt were made. The value for the flux obtained by this method was used for preliminary calculations of burnup and irradiation temperature.

The capsules were irradiated in 2 adjacent columns in the same MTR X-basket. Approximately the first 10 per cent of the irradiation was in the fuel lattice and the remaining 90 per cent was in the beryllium reflector. Since there was only one specimen in each capsule, the resulting cylindrical geometry enabled the temperature gradients in the fuel and capsule to be calculated analytically.

The measurements made prior to irradiation were repeated, except that no unclad weights could be obtained on the clad specimens nor could the core and sleeve be weighed separately. Volume changes for specimens containing porous end closures were calculated from measurements of lengths and diameters.

In addition to these measurements, an extensive postirradiation metallographic examination was performed. A metallographic sample was cut through the cross section at the midplane of each specimen. The metallographic studies included examination for porosity, geometry and size of the central vent, and the general appearance of the fuel assembly from the central vent out through the stainless steel cladding.

In an attempt to correlate the fission product release from the clad specimens with the pore size in the porous end caps, the NaK from the

irradiation capsules was dissolved in n-butyl alcohol and samples of the alcohol-NaK mixture were submitted for radiochemical analysis of the Sr^{90}, Cs^{137} and Ce^{144} concentrations in the sodium.

To provide information for the computation of fuel burnup, 0.094-in.-thick wafers were cut from the center of 4 of the specimens for uranium isotopic analysis. These 4 specimens provided a calibration for the integrated neutron fluxes indicated by the aluminum-cobalt-manganese alloy flux monitors in each of the capsules. Since the 10 capsules under consideration were irradiated in 2 adjacent columns, 2 burnup samples were taken from each column. One sample was taken from the region of highest monitor-indicated flux and one from the top of the columns, where there was a large flux gradient over a short vertical distance. A mass spectrographic analysis for uranium-234, -235, -236, and -238 concentrations was performed for each of the 4 specimens. The information from these analyses was combined with that obtained by the pre-irradiation isotopic analysis for computation of fuel burnup. Fuel burnups were calculated as shown in Appendix A.

The mass spectrographic analysis also provided the information necessary for the calculation of the flux levels at which the specimens were irradiated. With knowledge of the flux level, the heat generation within the specimens and the corresponding temperature gradients in the fuel assembly were calculated. These specimens were irradiated at 3 widely different flux levels, and an account of their temperature history is contained in Appendix B.

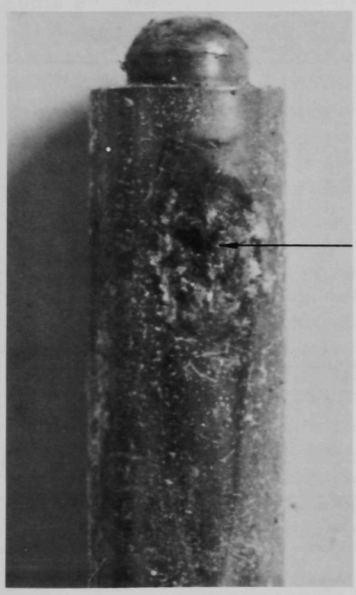
From the fuel geometry it is evident that the maximum temperatures reported existed over only a very small radial distance; 13 per cent of the total temperature drop in the entire irradiation capsule occurred within 0.031 in. in the enriched core, and another 25 per cent of the total temperature drop in the capsule occurred within 0.063 in. in the natural sleeve. Although the maximum temperatures reported for a few of the specimens seem doubtfully high, metallographic examination indicated that the temperatures reported are quite realistic.

In preparation for metallographic examination, the specimens were sectioned with an abrasive cutoff wheel and mounted in cold-setting plastic. A 10 per cent oxalic acid etch was used for studies on the stainless steel; an electrolytic etch of 4 parts phosphoric acid - 5 parts ethyl alcohol - 4 parts ethylene glycol was used for studies on the uranium-zirconium fuel alloy.

IRRADIATION DATA AND RESULTS

Two of the specimens (DF-1 and DF-10) were removed from the reactor after only 31.6 hr of irradiation, because of failure of the irradiation

capsules. The 2 columns of specimens were initially inserted inadvertently in a fuel lattice position where the maximum flux was almost 3 times that required to provide the maximum fuel temperatures of interest to this experiment. As a result, the walls of the irradiation capsules failed because of the high rate of generation of heat. Figure 9 shows the region where failure occurred in zirconium irradiation capsule DF-1. The area that failed was believed to be in contact with a capsule in the adjacent column and therefore was not adequately cooled by the reactor process water. The failure appears to have resulted from inadequate cooling rather than from excessive heat generation, since the 2 specimens DF-4 and DF-5 had higher internal heat generation than did specimens DF-1 and DF-10 and yet these capsules did not fail. However, specimens DF-4, DF-5, DF-8, and DF-9, which had approximately the same heat generation as specimens DF-1 and DF-10, were welded to the inside walls of their capsules and could not be removed from the capsules. Specimens DF-8 and DF-9 were adjacent to specimens DF-1 and DF-10 during the irradiation, and specimen DF-4 was adjacent to specimen DF-5. The areas where the fuel was welded to the capsule wall or where failure of the capsule wall occurred were probably the areas where the capsules were in contact.



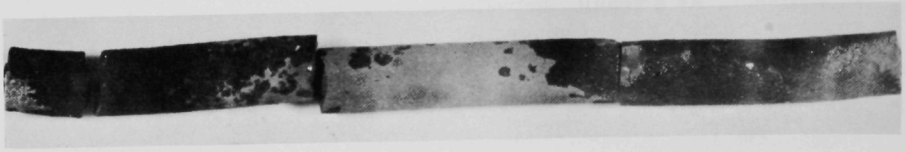
21930 3X

Figure 9. Area of Failure in Zirconium Irradiation Capsule DF-1.

Specimen DF-10 was in a lower flux than specimen DF-1 and could be removed from the capsule, even though it was severely damaged. The swollen fuel had exerted stresses sufficient to rupture the cladding in several places, and there were other regions where the cladding was at the point of incipient failure.

Two specimens (DF-12 and DF-14) were irradiated at relatively low burnups and temperatures. Specimen DF-12 was irradiated to a burnup of 2.7 a/o of the enriched core at a maximum fuel temperature of 190°C. Specimen DF-14 was irradiated to a burnup of 4.2 a/o of the enriched core at a maximum fuel temperature of 300°C. Upon removal of the cladding from specimen DF-12, the specimen broke into 4 pieces, as shown in Figure 10. The enriched core had undergone a slight amount of growth so that it protruded beyond the end of the sleeve a detectable amount at each end of the specimen. The cladding could not be removed from specimen DF-14. The

central vent was closed up, the enriched core was no longer cylindrical, and the fuel occupied almost the full internal volume of the stainless steel cladding.



23357 Figure 10. Specimen DF-12 after Removal of Cladding. 2X

The remaining 10 specimens were irradiated to higher burnups and temperatures, and were therefore of greater interest. Specimens representing the typical postirradiation appearance of these 10 specimens will be discussed. A summary of the irradiation data concerning the general appearance and physical measurements on the 10 specimens is contained in Table II. A summary of the information obtained from the metallographic examination is contained in Table III.

Table II

SUMMARY OF IRRADIATION INFORMATION OBTAINED WITH RESTRAINED AND VENTED U-2 w/o Zr ALLOY SPECIMENS

Specimen No.	Pore Size in End Caps, μ	Core Burnup, a/o	Maximum[a] Fuel Temp, °C	Cladding[a] Surface Temp, °C	Length Change, %	Diameter Change, %	Volume[b] Increase, %	% ΔV / a/o Burnup	Comments
DF-2	65	11.8	630	320	0	0	0	0	Clad specimen - very good, fuel swollen to cladding. NaK in axial hole.
DF-3	20	11.8	630	320	0	0	0	0	Clad specimen - very good, fuel swollen to cladding. NaK in axial hole.
DF-4	20	12.8	760	380	(c)	(c)	(c)	(e)	Unable to remove specimen from capsule.
DF-5	5	11.8	630	320	(c)	(c)	(c)	(e)	Unable to remove specimen from capsule.
DF-6	5	12.6	710	360	0	0	0	0	Eutectic formation near top of specimen.
DF-7	Solid	6.9	280	150	0	0	0	0	Fuel swollen to volume of cladding.
DF-8	Unclad	11.8	630	Unclad	(c)	(c)	(c)	(e)	Specimen broke into 3 pieces upon removal from capsule. NaK in axial hole of each piece.
DF-9	Solid	9.5	430	230	0[d]	0[d]	0[d]	0[d]	Specimen off center in capsule and bent, welded to capsule at a point near bottom of specimen.
DF-11	Unclad	6.9	280	Unclad	18	15	37	5.3	Specimen bent and surface irregular.
DF-13	Top - 0.005-in. hole Bottom - 0.010-in. hole	9.8	430	230	0.6	0.4	1.2	0.1	Fuel swollen to volume of cladding.

[a] The temperatures listed are those which existed at the beginning of the final 90% of the irradiation. A temperature-history of these specimens is contained in Appendix B.
[b] Computed on the basis of measurements of lengths and diameters.
[c] Unable to remove specimen from irradiation capsule.
[d] Except for area where fuel was welded to the capsule wall.
[e] Specimen restrained by irradiation capsule after expanding to the capsule walls.

Table III

SUMMARY OF METALLOGRAPHIC INFORMATION OBTAINED WITH RESTRAINED AND VENTED U-2 w/o Zr FUEL ALLOY SPECIMENS

Specimen No.	Metallographic Observations
DF-2	Vent closed; core solid with small pores. Zirconium foil broken in some areas and bonded with fuel alloy. Slight zirconium alloying with stainless steel.
DF-3	Vent closed; core has few large holes. Zirconium foil broken in some areas, bonded to fuel alloy in some areas, to stainless steel in other areas.
DF-4	Vent closed; large holes and voids due to melting in core and sleeve. No zirconium foil present, apparently dissolved by fuel alloy. Stainless steel dissolved in some areas, allowing fuel to flow to the capsule wall.
DF-5	Vent enlarged to 0.188 in. in diameter. Some zirconium alloyed with fuel alloy. Stainless steel difficult to find.
DF-6	Vent partially closed; large holes and voids in core. Zirconium foil broken in some areas, bonded to fuel alloy in others. Slight stainless steel-zirconium reaction.
DF-6 Burnout	Eutectic formation between fuel alloy and stainless steel. Many radial cracks from zirconium foil to core, one crack through cladding at fused area.
DF-7	Vent closed; large holes and voids in core and core-sleeve interface.
DF-8	Vent closed; core solid with small voids; core and sleeve separated by a series of voids. Residual NaK in these voids.
DF-9	Vent closed; slight bonding of zirconium to fuel alloy. Zirconium foil broken in some areas with slight alloying with stainless steel.
DF-11	Vent closed; core solid with small voids. Large holes with residual NaK in fuel. Fuel alloy severely eroded on outer surface.
DF-13	Vent closed; core has very small voids. Zirconium foil broken in very small areas and bonded to fuel alloy. Slight zirconium-stainless steel alloying.

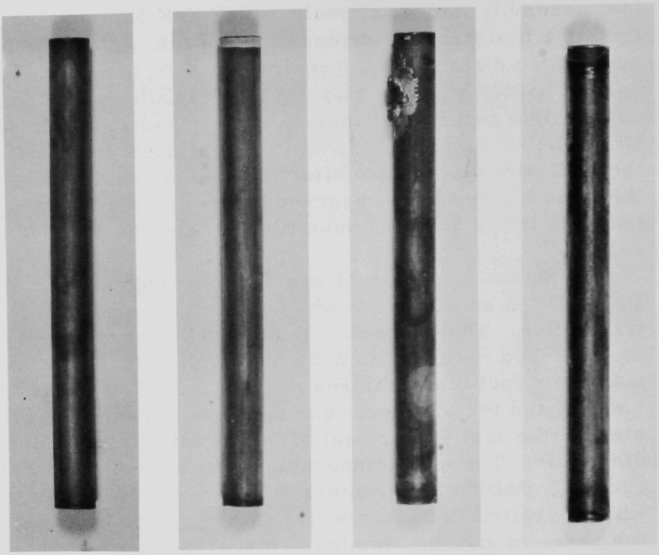
It was readily apparent, upon opening the irradiation capsules, that a significant volume of fission gases had been released from each vented fuel assembly into its capsule, since NaK squirted out of the capsule when the lathe tool first cut through the capsule wall. The NaK stream traveled an average distance of approximately 2 ft; this type of behavior had not been observed prior to this, even in capsules in which fuels had undergone appreciable volume expansion during irradiation. In specimens DF-2, DF-8, DF-9, and DF-13, a white, gelatinous precipitate was present in the NaK-alcohol mixture after the NaK-alcohol reaction was complete; this also had not been observed before. A part of this precipitate was included in the samples submitted for fission product analysis.

Shown in Figure 11 are 4 of the clad assemblies (specimens DF-2, DF-3, DF-6 and DF-7) in which no dimensional changes occurred during irradiation. These specimens achieved burnups from 6.9 to 12.6 a/o of the enriched core (1.2 to 2.2 a/o volume average of core and sleeve) at maximum fuel temperatures ranging from 280 to 710°C. The fuel temperatures listed for all specimens discussed are those which existed at the start of the last 90 per cent of the irradiation period, unless specified otherwise. The high temperature which specimen DF-6 experienced during cycle 89 enabled the uranium to dissolve a portion of the zirconium foil, which resulted in the formation of a region of low-melting eutectic between the uranium and the stainless steel jacket, as can be seen in Figure 11. Another clad specimen (DF-13) underwent an anomalous 1.2 per cent volume increase at a maximum fuel temperature of 430°C.

Figure 12 shows a cross section of specimen DF-3. The central vent is closed and the enriched core has been shifted toward one side of the specimen. Figure 13 shows a radial micro hardness traverse of specimen DF-3. A line of demarcation between the core and sleeve is evident. A zone of high porosity exists at the outer edge of the enriched core. Figure 14 shows the interface between enriched and natural alloy in specimen DF-6. The natural sleeve contains fine pores. The enriched core has many more and larger pores.

The stainless steel jackets of the 5 specimens (DF-2, DF-3, DF-6, DF-7, and DF-13) in which no swelling of the jacket occurred were cut open in an attempt to remove the fuel from the cladding. It was found that the fuel in all 5 of the clad assemblies had swollen to the full volume of the jackets and could not be separated from the cladding. In all specimens the central vent was completely closed.

The remaining 3 clad specimens (DF-4, DF-5, and DF-9) could not be removed intact from their irradiation capsules. They were swollen to the full inner diameter of the capsules or welded to the capsules in one or more locations. An increase of 16 per cent in the specimen diameter could be accommodated in the irradiation capsule. The central vents ranged from partially to completely open at each end of the specimens.



Negative No.	26601	26602	26607	26608
Specimen No.	DF-2	DF-3	DF-6	DF-7
Fuel Max. Temp, °C	630	630	710	280
Clad Surface Temp, °C	320	320	360	150
Core Burnup, a/o	11.8	11.8	12.6	6.9

Figure 11. Postirradiation Appearance of Clad Specimens in Which No Dimensional Changes Occurred During Irradiation. The Light-colored Region at the Top of Specimen DF-6 Was Due to a Reaction between the Uranium and the Stainless Steel (Magnification 1X).

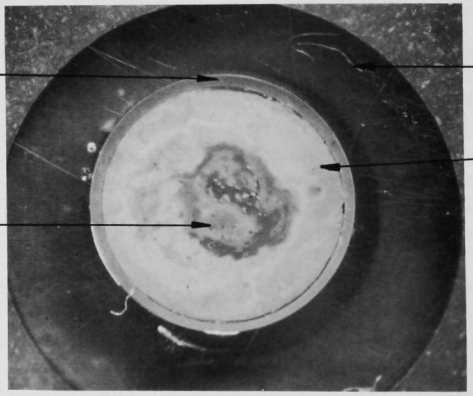
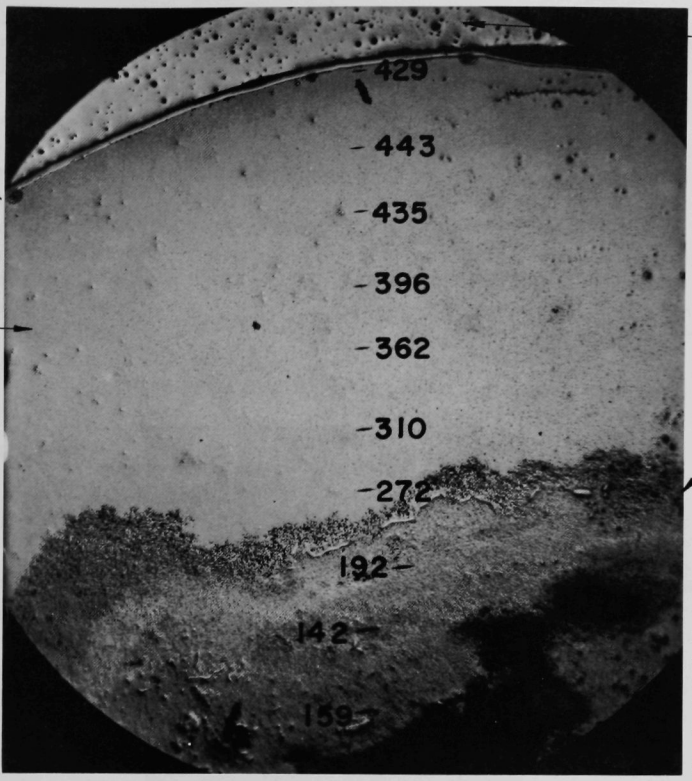


Figure 12

Cross Section of Specimen DF-3.

29257

6X

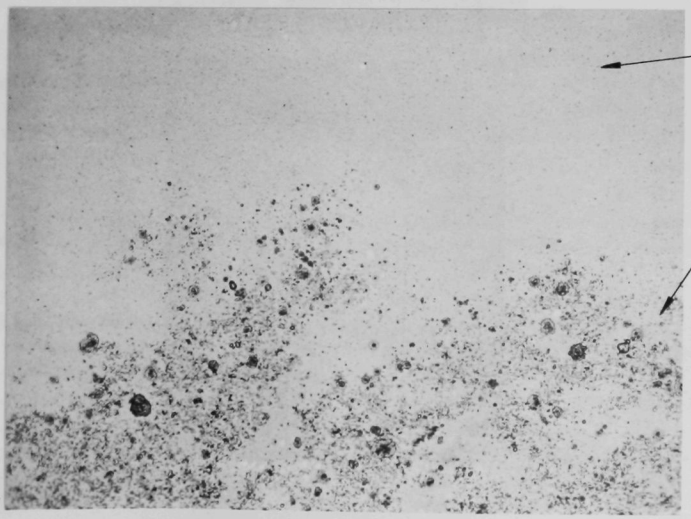


29256

50X

Figure 13

Knoop Hardness Profile from Core to Cladding in Specimen DF-3.



29267

250X

Figure 14

Interface of Enriched and Unenriched Areas in Specimen DF-6.

Figure 15 is a cross section of specimen DF-4 illustrating the melting and severe damage which is believed to have occurred during MTR cycle 89. Figure 16 is a cross section of specimen DF-5. The specimen had swollen to the full inner diameter of the irradiation capsule. The central vent had expanded proportionally. Four radial cracks extended from the central vent to the capsule wall. The stainless steel cladding, which was very obvious in specimens DF-3 and DF-4, could not be clearly seen. Figure 17 shows specimen DF-5 at higher magnification. A line of demarcation exists between the fuel and the cladding, although extensive alloying has evidently occurred between the fuel alloy and the stainless steel. There is a large crack extending from the central vent to the cladding. The etching characteristics appear to be a direct function of the burnup levels in the fuel. One would expect a higher burnup at the surface of the sleeve than within the sleeve and a much higher burnup in the enriched core, as is indicated in Figure 17.

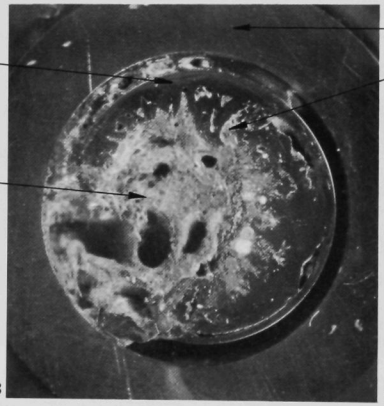


Figure 15

Cross Section of Specimen DF-4, Showing the Melting Which Is Believed to Have Occurred During Cycle 89.

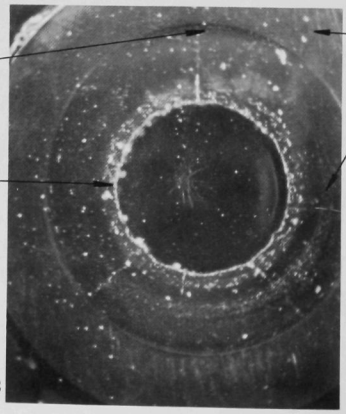
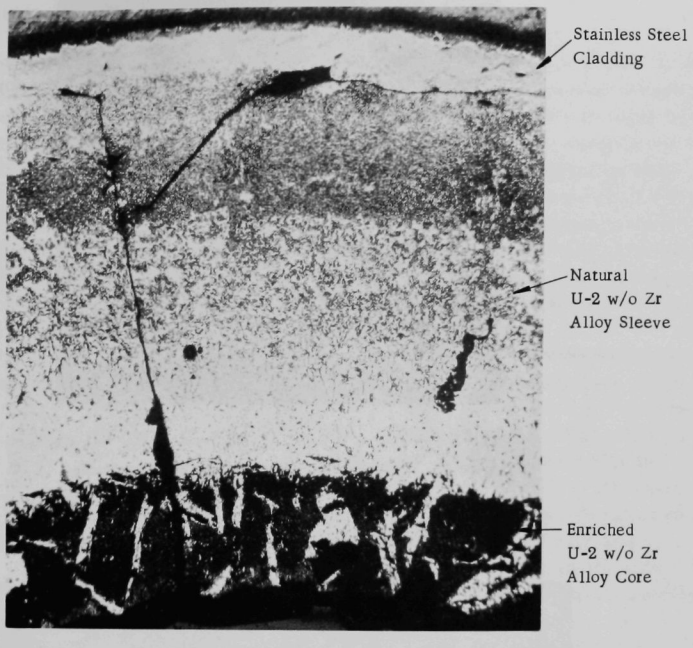


Figure 16

Cross Section of Specimen DF-5, Which Expanded to the Full Inner Diameter of the Capsule.

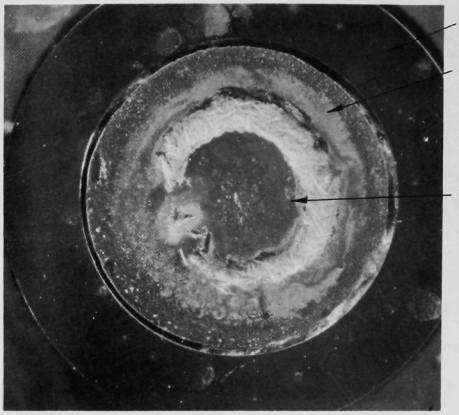


27050 50X

Figure 17. Microstructure of Specimen DF-5 after Etching with H_3PO_4.

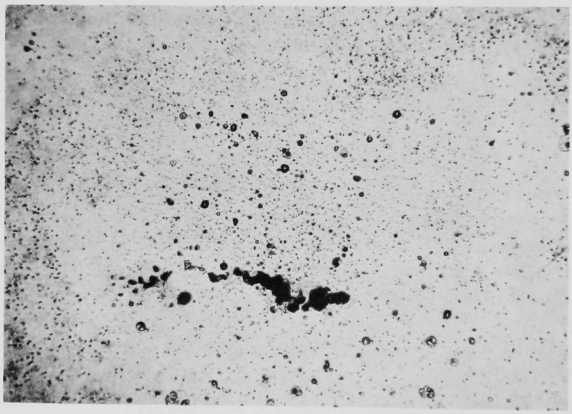
The 2 specimens DF-8 and DF-11 were unclad. Specimen DF-8 could not be removed from its irradiation capsule. Figure 18 is a cross section view of specimen DF-8. The central vent appears to be completely closed. The specimen is not swollen to the full inner diameter of the capsule. The raised white structure which almost completely surrounds the enriched core is oxidized NaK which oozed out of the interface between the core and the sleeve. Figure 19 shows the central vent area of specimen DF-8. The vent is almost completely filled with solid material. The central region has a few large voids and many smaller ones; the core around this contains many larger voids. Surrounding the core were many large voids which almost completely separated the core and sleeve at the longitudinal center of the specimen.

The other unclad specimen, DF-11, had undergone a 37 per cent volume increase after 6.9 a/o burnup of the core at 280°C. The specimen is shown in Figure 20. A diameter increase of 51 per cent could occur in an unclad specimen before restraint would develop from the irradiation capsule. The central vent in specimen DF-11 was closed at the top of the specimen and the enriched core protruded slightly beyond the sleeve, as can be seen in Figure 20.



29237 6X

Figure 18. Cross Section of Specimen DF-8, an Unclad Specimen, Showing that the Central Vent Is Completely Closed, and the Large Voids at the Core-sleeve Interface Are Filled with NaK.



29242 50X

Figure 19. Central Vent Area of Specimen DF-8, Showing the Remainder of Central Vent and Relatively Low Porosity.

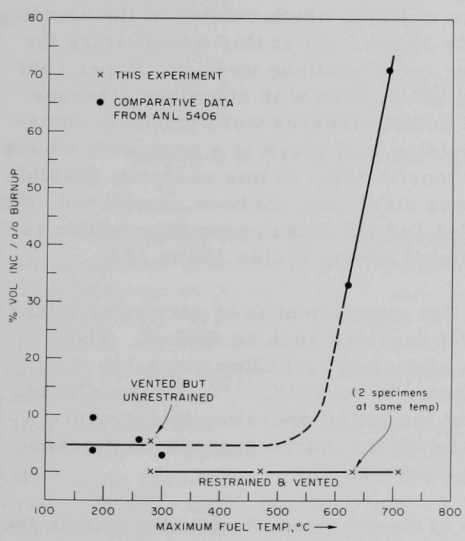
26612 1X

Figure 20. Postirradiation Appearance of Specimen DF-11, Which Has Undergone a 37 per cent Volume Increase after 6.9 a/o Burnup of the Core at 280°C.

DISCUSSION OF RESULTS

Previous work indicated that unrestrained and nonvented uranium-2 w/o zirconium alloy begins to swell catastrophically at fuel temperatures of approximately 500°C at burnup levels ranging from 0.18 to 5.3 total atom per cent.[9] In the present experiment, vented and clad fuel assemblies experienced no volume increases at maximum fuel temperatures in the range from 630 to 710°C to burnups of 6.9 to 11.8 a/o of the enriched core (2.1 to 2.2 a/o volume average). The damage to those specimens which could not be removed from their irradiation capsules is believed to have occurred during their initial high-flux cycle in the reactor. The damage was therefore primarily thermally induced rather than irradiation-induced.

Figure 21 illustrates swelling rates as a function of temperature for bare fuel and for restrained and vented fuel compared with data from reference 9 on unclad and unrestrained fuel alloy. The clad specimens which could not be removed from their irradiation capsules could not be measured for volume changes and are therefore not shown in Figure 21. The temperature at which swelling occurred in the clad specimens was apparently between 710 and 760°C. The one point available for vented but unclad fuel suggests that in this experiment the central vent by itself did not significantly reduce the swelling rate of the fuel.



106-5677-A

Figure 21. Effect of Irradiation Temperature on the Rate of Volume Increase in Uranium-2 w/o Zirconium Alloy.

In this experiment, some of the clad specimens experienced no measurable dimensional changes, another underwent a diameter increase of 0.2 per cent, and 2 underwent diameter expansions of 16 per cent. The specimens which experienced no measurable dimensional changes were obviously below the stress necessary to induce creep for the time which the cladding was at an elevated temperature. An examination of available pertinent creep data for Type 304 stainless steel indicates that a stress level below 14,000 psi would not cause a measurable amount of creep in the cladding of those specimens which had a long-time (3,630 hr) mean cladding temperature of approximately 330°C

(specimens DF-2, DF-3, and DF-5). In the specimen which had a total deformation of 0.2 per cent at a mean cladding temperature of approximately 250°C, the creep rate was 5×10^{-7} in./in.-hour. The approximate stress to cause this creep rate could not be calculated reliably, since published creep data were not located at temperatures as low as 250°C. However, a stress of approximately 35,000 psi would induce a creep rate of this magnitude at a temperature of 350°C. Since a reliable extrapolation of creep rate versus temperature data cannot be performed, the only valid information obtained with this specimen is that the stress level imposed on the cladding was greater than 35,000 psi.

For the 2 specimens which expanded to the full inner diameter of the capsules, it seems reasonable to assume that the deformation occurred during the first few hours of cycle 89. For this situation, short-time yield-point data would more closely approximate actual conditions than creep data. In a pure tensile test, yielding would occur at a stress of approximately 12,000 psi at the temperature of 650°C which existed in the cladding. Since the elongation is approximately 38 per cent at this temperature for Type 304 stainless steel, the yielding could continue up to the 16 per cent experienced by specimens DF-4 and DF-5, even with combined stresses and irradiation embrittlement. The actual stresses were probably somewhat lower than 12,000 psi, since yielding took place at a somewhat slower rate than that utilized in a standard tensile test. In this analysis, possible early radiation effects on the stainless steel have not been considered. For all specimens except specimens DF-4 and DF-5 the creep information is based on the temperatures which existed during cycles 103 to 112.

The information obtained in this experiment is of particular interest to fuel element programs for fast reactors such as EBR-II. The second core of EBR-II will utilize a plutonium fuel alloy which has a swelling temperature in the range from 350 to 400°C.[12] The results of the present experiments indicate that the useful operating temperature range of this fuel could be extended several hundred degrees Centigrade by utilization of the proper cladding material.

Data obtained on the release of fission products into the NaK in the irradiation capsule is shown in Table IV. The fission product concentration outside the cladding appeared to be a direct function of irradiation temperature rather than of the pore size of the end caps. The only exception to the increase in fission product release with increased irradiation temperature was for specimen DF-4. This exception is probably because of the melting which occurred in DF-4 during cycle 89. Upon melting, the original geometry and the integrity of the fuel were lost, a condition which would affect the fission product release of this specimen.

Table IV

FISSION PRODUCT RELEASE FOR RESTRAINED AND VENTED U-2 w/o Zr FUEL ALLOY SPECIMENS

Specimen No.	Max Fuel Temp, °C	Core Burnup, a/o	Pore Size in End Caps, μ	Activity[a]				
				Ce^{144}	Sr^{90}	Cs^{137}	$\frac{Sr^{90}}{Ce^{144}}$	$\frac{Cs^{137}}{Ce^{144}}$
DF-2	630	12.0	65	14	18,200	35,500	1300	2,540
DF-3	630	12.0	20	95	33,100	60,100	348	634
DF-4	760	13.0	20	60	591	2,100	10	35
DF-5	630	12.0	5	17	532	13,000	31	765
DF-6	710	12.8	5	312	85,000	112,000	273	356
DF-7	280	7.0	Solid	1	427	16,300	427	16,300
DF-8	630	12.0	Bare	3,330	111,000	72,600	33	22
DF-9	470	9.6	Solid	107	12,500	58,000	117	540
DF-11	280	7.6	Bare	169	9,640	35,000	57	210
DF-13	430	9.9	Top 0.005 in. Bottom 0.010 in.	34	17,900	57,600	526	1,715

[a] Activity is in disintegrations per sec per μg of sodium.

From the following simple analysis one might expect roughly an equal number of atoms of each isotope to be released into the NaK coolant. The diameter of the Sr^{90}, Cs^{137}, and Ce^{144} atoms are 4.30, 5.24, and 3.64 Å, respectively. The pores provided for fission product release were from 10,000 to 130,000 times larger in diameter than the diameters of these atoms, so that, neglecting surface absorption effects, all 3 atoms could move through the pores easily. The half-life of Cs^{137} is 40 times longer than the half-life of Ce^{144}, and the half-life of Sr^{90} is 36 times longer than that of Ce^{144}. The half-life of Cs^{137} is only 1.11 times longer than that of Sr^{90}; therefore, they are considered to be the same for this case. The fission yields of Sr^{90}, Cs^{137}, and Ce^{144} are 5.77, 6.00, and 6.15 per cent, respectively. Hence, they are considered as equivalent within the range of experimental error associated with this experiment. Finally, the neutron-capture cross section of Sr^{90} is one barn, of Cs^{137} less than 2 b, and of Ce^{144} 1.3 b, so again they are considered equivalent. Taking all these factors into account, on the day the samples were counted the Ce^{144} activity should have been higher than the Cs^{137} activity by a factor of 6 and higher than the Sr^{90} activity by a factor of 5. As shown in Table IV, this condition did not exist. A possible reason for this difference between the activity expected and that obtained may be a preferential solubility of these 3 elements in NaK or a preferential plating of these elements on the capsule wall.

Two experimental conditions which further complicate interpretation of the data are as follows. One may recall that between each porous end cap and the fuel there was a diffusion barrier of 0.001-in.-thick zirconium foil. For an atom to get out of the fuel assembly, the atom had to go around the foil before reaching the porous end cap. It is doubtful that the geometry of the foil was identical in each assembly. Also, during irradiation the fuel swelled to the full volume of the cladding in each of the clad specimens. This pushed the foil against the stainless steel end cap which closed up the holes in the end cap.

CONCLUSIONS

1. Type 304 stainless steel cladding, 0.010 in. thick, restrained swelling in vented, 0.22-in.-diameter, uranium-2 w/o zirconium alloy fuel specimens to burnup levels of the enriched core of 12.6 a/o and at maximum fuel temperatures up to 710°C. Hence, the maximum operating temperature of the fuel was extended at least 200°C, and the burnup level extended several atom per cent.

2. A central vent alone did not appear to provide any significant additional dimensional stability to this fuel alloy.

3. Fission product release was predominantly a function of fuel temperature rather than the pore size in the vented end caps.

ACKNOWLEDGMENTS

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APPENDIX A

BURNUP COMPUTATIONS

Because of the high burnups to which these specimens were irradiated and because of the unique geometry of the fuel, the following presentation of the methods by which the burnups were computed is included.

A mass spectrographic analysis for uranium isotopes was performed on the inner core and the outer sleeve prior to the assembling of the 2 sections into a duplex specimen. Because of the diffusion, swelling, melting, and distortion that occurred during the irradiation, the cores and sleeves could not be separated to obtain a postirradiation uranium isotope concentration of the core and sleeve separately. Therefore, the $\frac{1}{16}$-in.-thick cross-sectional wafer cut from the longitudinal center of the 4 specimens included both the core and sleeve. Hence, the postirradiation uranium isotope concentration obtained was the volume average for the duplex assembly.

From the preirradiation isotopic analysis, the volume-averaged uranium isotope concentration was computed for the duplex assemblies prior to irradiation. The depletion of U^{235} was then computed by several different methods. The first method used was to consider that the weight of U^{238} did not change during irradiation, since its conversion to plutonium is much slower than the rate of depletion of U^{235}. Work at Argonne National Laboratory[9] and at the Phillips Petroleum Company[13] has shown that this assumption leads to less than a one per cent error in the final results, since at least 95 per cent of the uranium lost is by fission. The weight of U^{235} present in the samples before and after irradiation was obtained, and the depletion of U^{235} is represented by the difference. From this value, the per cent of U^{235} fissioned was readily calculated, with the aid of the fission and absorption cross sections from reference 14. The uranium burnup and total atom per cent burnup were computed from this value. The burnup of specimen DF-4 is presented as an example.

Table AI

URANIUM ISOTOPE CONCENTRATION IN DUPLEX ASSEMBLY

	U^{234}	U^{235}	U^{236}	U^{238}
Preirradiation, w/o	0.037	3.313	few ppm	96.648
Postirradiation, w/o	0.021	0.593	0.287	99.099

Hence - for constant U^{238} weight:

(Wt of U before irrad.)(% U^{238} before irrad.) = (Wt of U after irrad.)(% U^{238} after irrad.)

$$\text{Wt of U after irrad.} = \frac{(\text{Wt before irrad.})(\%\ U^{238}\ \text{before irrad.})}{(\%\ U^{238}\ \text{after irrad.})}$$

$$31.442\ \text{gm} \times \frac{0.9665}{0.9910} = 30.665\ \text{gm} \quad .$$

Thus the masses of U^{235} before and after irradiation were

$$\text{gm } U^{235}\ \text{before} = 0.03313 \times 31.442 = 1.042$$

$$\text{gm } U^{235}\ \text{after} = 0.00593 \times 30.665 = 0.182$$

$$\text{gm } U^{235}\ \text{lost} = 0.860$$

$$\%\ U^{235}\ \text{depletion} = \frac{1.042 - 0.182}{1.042} \times 100 = 82.55\%$$

$$\%\ U^{235}\ \text{fissioned} = \frac{\sigma_f}{\sigma_a}(\text{depletion}) = \frac{582 \times 10^{-24}\ \text{cm}^2}{694 \times 10^{-24}\ \text{cm}^2}(82.55)$$

$$= 0.839 \times 82.55 = 69.26 \quad ,$$

where

σ_f = fission cross section (582×10^{-24} cm^2)

σ_a = absorption cross section (694×10^{-24} cm^2) .

Uranium burnup = (% fissioned)(% U^{235} in uranium)

$69.26 \times 0.03313 = 2.29$ a/o .

U-2 w/o Zr alloy contains 94.92 a/o U .

Hence: Average total atom per cent burnup = (uranium burnup)(a/o uranium in alloy)

Average total atom per cent burnup = $2.29 \times 0.9492 = 2.17$.

Therefore,

Average burnup = 2.2 a/o .

The second method employed was based on a simple algebra involving the per cent U^{235} before and after irradiation as follows. This method neglects the apparent increase in U^{238}.

From Table AI

$$\% \ U^{235} \text{ depletion} = \left(\frac{\% \ U^{235} \text{ before} - \% \ U^{235} \text{ after}}{\% \ U^{235} \text{ before}} \right) \times 100$$

$$\left(\frac{0.03313 - 0.00593}{0.03313} \right) \times 100 = 82.10\% \ .$$

$$\% \ U^{235} \text{ fissioned} = \frac{\sigma_f}{\sigma_a} \times 82.1 = \frac{582}{694} \times 82.1 = 68.88 \ .$$

U burnup = 68.88 x 0.03313 = 2.28 a/o .

Average total atom per cent burnup = 2.28 x .9492 = 2.16 .

Therefore,

Average burnup = 2.2 a/o

The third method used to compute the burnup was equation 5 of ANL-5406 (reference 9). There is a typographical error in the equation as it appears in the reference. It should read as follows:

$$\text{Total a/o burnup} = (\text{a/o U in specimen}) \left[\frac{(a_0 + b_0) - (a + b)}{(1 + \alpha) - (a + b)} \right] \times 100 \ ,$$

where

a_0 = fraction of uranium that is U^{234} before irradiation

a = fraction of uranium that is U^{234} after irradiation

b_0 = fraction of uranium that is U^{235} before irradiation

b = fraction of uranium that is U^{235} after irradiation

$\alpha = \sigma_c / \sigma_f$

and

σ_c = capture cross section (112×10^{-24} cm^2).

$$\text{Total a/o bu} = 0.9492 \left[\frac{(0.00037 + 0.03313) - (0.00021 + 0.00593)}{1.19 - (0.0002 + 0.00593)} \right] \times 100$$

Average burnup = 2.2 a/o

To compute the burnup in the enriched core, one could reason that if 82.55 per cent of the U^{235} were depleted in the duplex assembly the same amount would be depleted in the core, since the burnout rate in the core is the same as in the assembly. Also, a little more than 80 per cent of the U^{235} present is in the enriched core. By this analysis for DF-4 one would obtain as follows:

Preirradiation U^{235} content of core = 19.3%

% U^{235} depletion = 82.55%

% U^{235} fissioned = 82.55 x 0.839 = 69.26

Total U burnup = 69.26 x 0.193 = 13.37%

Total burnup = 13.37 x 0.9492 = 12.7 a/o .

Therefore,

Core burnup = 12.7 a/o .

However, this method alone was not employed, but the 3 methods previously described were also used, and the results were in good agreement. A method which is effectively the same as the first one was based on the use of the mass of U^{235} in the core before and after irradiation. A simple ratio was set up to obtain the mass of U^{235} in the core after irradiation as follows:

$$\frac{\text{Mass } U^{235} \text{ in core before irrad.}}{\text{Mass } U^{235} \text{ in duplex before irrad.}} = \frac{\text{Mass } U^{235} \text{ in core after irrad.}}{\text{Mass } U^{235} \text{ in duplex after irrad.}}$$

For DF-4 this gave

$$\frac{0.849}{1.042} = \frac{\text{Mass } U^{235} \text{ in core after irrad.}}{0.182}$$

or

Mass U^{235} in core after = (0.849)(0.182)/1.042 = 0.148 gm .

Hence, the % depletion of U^{235} is

$$\% \, U^{235} \text{ depletion} = \left(\frac{0.849 - 0.148}{0.849} \right) \times 100 = \frac{70.1}{0.849} = 82.57$$

$$\% \, U^{235} \text{ fissioned} = 82.57 \times 0.839 = 69.28$$

$$\% \text{ U burnup} = 69.28 \times 0.193 = 13.37 \quad .$$

Therefore,

$$\text{Core burnup} = 13.37 \times 0.9492 = 12.7 \text{ a/o} \quad .$$

The same ratio method was used to obtain the fractions of U^{234} and U^{235} in the core for use in equation 5 of ANL-5406. The example for DF-4 by this method is:

$$\text{Total a/o bu} = 0.9492 \left[\frac{(0.00235 + 0.193) - (0.00133 + 0.0345)}{1.19 - (0.00133 + 0.0345)} \right] \times 100$$

and

$$\text{Core burnup} = 13.2 \text{ a/o} \quad .$$

The use of simple algebra led to a burnup value of 12.8 total atom per cent. Table AII contains the results of the burnup computations by the various methods. From these values, the perturbed fluxes necessary to induce the average burnups were computed by use of the standard burnup equation:

$$\text{Total a/o burnup} = Af (\sigma_f/\sigma_a)[1 - \exp(-\phi t \sigma_a)] \quad ,$$

where

A = atomic per cent uranium in fuel

f = fraction of uranium which is U^{235}

ϕ = neutron flux (neutrons/cm^2-sec)

t = irradiation time (sec).

From the results obtained from these 4 specimens, the average perturbed fluxes incident upon the other 6 specimens were obtained from a normalized flux plot, using the cobalt monitors contained in each capsule. The fluxes obtained by this method were used to compute the burnup in the remaining 6 specimens. The flux and burnup data for all 10 specimens are shown in Table AIII.

Table AII

BURNUP IN VENTED AND CLAD FUEL SPECIMENS COMPUTED BY VARIOUS METHODS

Specimen No.	Total Atom Per Cent Burnup							
	Enriched Core				Duplex Assembly			
	By Algebra	Same U^{238} Weight	Eq. 5 of ANL-5406	Average	By Algebra	Same U^{238} Weight	Eq. 5 of ANL-5406	Average
DF-4	12.6	12.7	13.2	12.8	2.2	2.2	2.2	2.2
DF-6	12.4	12.5	13.0	12.6	2.2	2.2	2.2	2.2
DF-9	9.2	9.3	9.9	9.5	1.7	1.7	1.7	1.7
DF-13	9.5	9.6	10.2	9.8	1.7	1.7	1.7	1.7

Table AIII

BURNUP IN VENTED AND CLAD FUEL SPECIMENS

Specimen No.	Total Atom Per Cent Burnup		
	Enriched Core	Natural Sleeve	Duplex Assembly
DF-2	11.8	0.5	2.1
DF-3	11.8	0.5	2.1
DF-4	12.8	0.5	2.2
DF-5	11.8	0.5	2.2
DF-6	12.6	0.5	2.2
DF-7	6.9	0.3	1.2
DF-8	11.8	0.5	2.0
DF-9	9.5	0.4	1.7
DF-11	6.9	0.3	1.2
DF-13	9.8	0.4	1.7

APPENDIX B

TEMPERATURE COMPUTATIONS

From the neutron fluxes required to produce the burnups obtained in the specimens, the power generation and, subsequently, the temperature rises in the fuel assemblies could be computed. The neutron flux obtained from the burnup computations is the average flux incident upon the specimens during the irradiation. Use of this flux for computing the temperature rises in the fuel would give the time-averaged irradiation temperature of the fuel and not the maximum. This situation exists because the specimens were located in 3 different positions in the MTR during their irradiation.

The specimens were irradiated in lattice position L54 for 31.6 hr during cycle 89 at a reported unperturbed flux of 5.5×10^{14} neutrons/cm^2-sec. They were then removed from the reactor, because of failure of capsules DF-1 and DF-10, and reinserted for 343.2 hr during cycle 93 in lattice position L58 at a reported unperturbed flux of 4.2×10^{14} neutrons/cm^2-sec. The final 94 per cent of the irradiation time (3,629 hr during cycles 103-112) was in reflector position A38SE at a reported unperturbed flux of 3.2×10^{14} neutrons/cm^2-sec. Even though the unperturbed fluxes reported by the MTR are usually somewhat higher than neutron flux monitors and subsequent burnup analyses indicate, one can readily appreciate that the major portion of the temperature damage to the fuel occurred during the first 31.6 hr of irradiation. Because of this, it is necessary to know the temperatures which existed in the specimens during the first few hours of irradiation.

The average perturbed flux on DF-4 obtained from the burnup computations was 1.8×10^{14} neutrons/cm^2-sec; the identical value was obtained by calculating a time-weighted average of the reported unperturbed fluxes to which DF-4 was exposed and by applying the Brad Lewis method to these fluxes.[15] Thus, the MTR-reported fluxes can be used to calculate the approximate temperatures of the fuel during the irradiation in the 3 different positions; these temperatures are significantly different than the temperatures obtained from the average flux necessary to produce the burnups obtained. Although this type of analysis necessarily contains certain assumptions, it appears to be the best method available to obtain a knowledge of the maximum temperatures experienced by the specimens.

The heat production in the specimens was calculated by the method presented by Glasstone.[16] Since the heat production in the core was greater by a factor of 22 than the heat production in the sleeve, only the heat generation in the core was considered for the temperature computations. Also, since the maximum difference in U^{235} weight per core section

from the mean value was 5 per cent, the mean weight of U^{235} per core section was employed. The temperature drops in the fuel and the capsule were computed by standard, annular cylindrical heat transfer methods as presented by Bonilla.[17] The following assumptions were utilized in the temperature analysis:

1. All heat flow was radial and was by conduction.

2. There were 180 Mev of energy released in the fuel per fission event.[18]

3. No adjustment was made for U^{235} depletion, change in thermal conductivities, or change in perturbation factor as the irradiation progressed.

4. All heat generation was in the core.

5. The temperature drop in the zirconium foil was insignificant.

6. There is no film drop between the NaK and the materials in the capsule (i.e., complete wetting occurred).

7. The specimen was in the radial center of the capsule.

The maximum temperatures experienced by the specimens during their irradiation in the 3 different positions are given in Tables BI, BII, and BIII of this Appendix. The temperatures reported are based on the preirradiation geometry of the fuel assembly. In 2 of the specimens, the original geometry was lost during the first cycle, during which melting of the center portion of the fuel occurred. In all of the specimens, distortion occurred which also changed the fuel geometry.

Table BI

CALCULATED MAXIMUM TEMPERATURES (°C) IN FUEL ASSEMBLIES DURING CYCLE 89

Specimen No.	Core Center	Core Surface	Sleeve Surface	Cladding Inner Surface	Cladding Outer Surface
DF-1	1180	1030	740	600	580
DF-2	430	380	280	240	230
DF-3	1010	880	640	520	500
DF-4	1300	1140	820	670	640
DF-5	1300	1140	820	670	640
DF-6	1010	880	640	520	500
DF-7	460	400	300	250	240
DF-8	1180	1030	740	Unclad	Unclad
DF-9	800	700	510	420	400
DF-10	800	700	510	420	400
DF-11	430	380	280	Unclad	Unclad
DF-13	460	400	300	250	240

During the early part of cycle 89, almost the entire core section at the longitudinal center of specimen DF-4 melted; this resulted in a redistribution of the U^{235} atoms and closing of m

Table BII

CALCULATED MAXIMUM TEMPERATURES (°C) IN FUEL ASSEMBLIES DURING CYCLE 93

Specimen No.	Core Center	Core Surface	Sleeve Surface	Cladding Inner Surface	Cladding Outer Surface
DF-2	850	740	540	440	420
DF-3	850	740	540	440	420
DF-4	1010	880	640	520	500
DF-5	850	740	540	440	420
DF-6	950	830	600	490	470
DF-7	360	320	240	200	190
DF-8	850	740	540	Unclad	Unclad
DF-9	570	500	360	300	290
DF-11	360	320	240	Unclad	Unclad
DF-13	570	500	370	300	290

Table BIII

CALCULATED MAXIMUM TEMPERATURES (°C) IN FUEL ASSEMBLIES DURING CYCLES 103 TO 112

Specimen No.	Core Center	Core Surface	Sleeve Surface	Cladding Inner Surface	Cladding Outer Surface
DF-2	630	560	410	340	320
DF-3	630	560	410	340	320
DF-4	760	660	480	400	380
DF-5	630	560	410	340	320
DF-6	710	630	460	370	360
DF-7	280	250	190	160	150
DF-8	630	560	410	Unclad	Unclad
DF-9	430	380	280	240	230
DF-11	280	250	190	Unclad	Unclad
DF-13	430	380	280	240	230

Also, at temperatures near the melting point, diffusion occurs rapidly, which could have resulted in a distribution of the U^{235} atoms over a larger volume than the region which melted. With the U^{235} atoms distributed over a larger volume, the heat production per unit volume was decreased and resulted in lower temperatures throughout the fuel and capsule. An additional factor which would tend to lower the temperature is that the perturbation factor increased upon closing of the central vent, since this factor is inversely proportional to the surface area of the fuel. This resulted in a lower fission rate in the fuel. Since the maximum temperatures are the temperatures of prime interest, a detailed analysis of the lower temperatures is not included. It seems reasonable to assume that the geometry that existed after cycle 89 remained approximately the same for the remainder of the irradiation. Hence, the temperatures for cycles 93 and 103 to 112 are affected primarily by U^{235} depletion and the diffusion of U^{235} into the outer sleeve.